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CONTRIBUTIONS FROM THE JEFFERSON PHYSICAL LABORATORY, HARVARD UNIVERSITY.

FRICTION IN GASES AT LOW PRESSURES.

By J. L. Hogg.

Presented by John Trowbridge June 29, 1909; received June 29, 1909.

Under the title "Friction and Force due to Transpiration as dependent on Pressure in Gases," there was published [1] some time ago an account of some experiments made to determine the relation between the friction of a gas and the pressure in it, and also the relation between the force exerted by a lamp on a mica vane, blackened on the face which is turned towards the lamp, and the mean pressure in the gas in which the vane is placed. The three-fold purpose of the investigation was pointed out there, viz.:

First, to investigate the relation between friction and pressure where the pressures were so small that "slip" is appreciable; second, to determine the relation of transpiration force in the special form of apparatus described there; [2] and, third, to make use, if possible, of these two relations to test the validity of the McLeod gauge measurements of pressure, and, if these measurements should prove unreliable, to make use of one of the relations named above to measure gas pressure.

There has been much delay in carrying out the investigation with the apparatus improved in the manner indicated in the closing paragraphs of that paper, but now some results have been obtained in so far as the friction problem is concerned.

As was pointed out in the paper mentioned, the investigation was defective in two respects. It was found that, in spite of the care which was taken to exclude mercury vapor from the apparatus, some of this vapor was undoubtedly present. This no doubt was due to the fact that the whole apparatus had to be maintained at a high temperature for long periods to insure drying, and thus the presence of the least speck of liquid mercury would cause, when evaporation took place, the diffusion of comparatively large quantities of the vapor through the apparatus. Again, the logarithmic decrement due to the friction in the suspending fibre was not determined directly by experiment, and in the discussion of the results obtained its value was calculated.

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[1] Proc. Am. Acad., **42**, 6 (1906).

[2] See p. 129 of that paper.

The details of the method since used to exclude the mercury vapor and to determine the decrement due to friction in the fibre will appear later. Meanwhile a summary of what has been accomplished is given here.

First, the decrement due to the friction in the suspending fibre of the viscosity apparatus has been determined experimentally.

Second, mercury vapor has been excluded to such a degree that, even when the whole apparatus, in which the presence of the vapor would be objectionable, was kept at a temperature of 150° C., the mercury lines were absent from the spectrum of the gas enclosed.

Third, the value of the decrement has been obtained for hydrogen over a range of pressures extending from atmospheric pressure to 0.000016 mm. as indicated by the McLeod gauge.

Fourth, an equation relating pressure to decrement has been obtained which applies well at all pressures below 0.1 mm. as far as pressures have been measured. The equation, above mentioned, is of the form of Sutherland's equation given tentatively in the former paper. It is

$$\left(\frac{k}{l-\mu}-1\right)p=c.$$

In this equation k and c are constants to be determined from the observations; l is the decrement due to whatever friction there is in the gas under examination and to the friction in the fibre; p is pressure; μ is the decrement due to the friction in the fibre. Its value has been measured directly. The significance of the two slightly differing values of μ, namely, $\mu = 0.000020$ and $\mu = 0.000022$, which are found in the following table, will appear later when the measurement is discussed in detail. The first column of the table contains a series of values of the decrement for hydrogen, each of which corresponds to a definite pressure in the gas. The various values of the pressure are given in the second column. The first three of them were obtained from a manometer. Those which are marked thus,*, were obtained from measurements made with the McLeod gauge, while the others were obtained from a curve plotted from the directly observed values of the decrement and pressure. From two values of p, the corresponding values of l, and the value of μ, there are obtained two equations for the determination of the constants k and c in the above equation. These determined, it is clear that from any value of l, within the range

TABLE I.

HYDROGEN.

Log. Dec.	p (observed).	p (calculated) $\mu = 0.000020$.	p (calculated) $\mu = 0.000022$.
0.07942	760.0	......	......
0.07937	435.0	......	......
0.07927	103.0	......	......
0.07768*	8.89*	......	......
0.06902*	1.24*	......	......
0.05423*	0.410*	......	......
0.02861*	0.105*	0.109	0.109
0.01140	0.0300	0.0302	0.0302
0.01056	0.0275	0.0276	0.0275
0.00936*	0.0239*	......	......
0.00887	0.0225	0.0225	0.0225
0.00710	0.0175	0.0174	0.0174
0.00620	0.0150	......	......
0.00525	0.0125	0.0125	0.0125
0.00434*	0.0102*	0.0102	0.0102
0.00426	0.0100	0.00998	0.00998
0.00306*	0.00704*	0.00702	0.00702
0.00220	0.00500	0.00497	0.00496
0.00112	0.00250	0.00247	0.00246
0.000459*	0.00098*	0.00097	0.00097
0.000215*	0.00042*	0.00043	0.00043
0.000029*	0.000016*	0.000020	0.000015

indicated above, the corresponding value of p can be obtained from the formula by a simple calculation. The numbers thus obtained for the various values of l are given in the third and fourth columns.

It would, therefore, seem highly probable that so far as hydrogen is concerned the McLeod gauge can be relied upon for pressures as low as the lowest used, and which are recorded in Table I; and that, in the case of hydrogen, the measurement of friction can be used as a convenient and accurate method of measuring pressure, provided care is taken to exclude mercury vapor. This matter will be discussed at length later.

The details of the methods used to overcome the difficulties named above follow:

Measurement of Decrement due to the Friction in the Fibre.

Referring to Figure 1 it will be seen that the tube C is inserted in such a position that nothing can pass to the viscosity apparatus from the McLeod gauge, B, or from the pump, which is connected to D, without passing through it. This tube, C, therefore, replaces the tubes of sulphur and silver whose purpose was explained in the earlier paper. C is filled with granular charcoal, and is so arranged that either a cylindrical electrical heater or a long Dewar vessel can enclose it. When C had been placed in position and sealed in place, the whole apparatus was exhausted through D by means of the mechanical pump, and then dry air was allowed to pass in through an opening placed near the pump. The exhaustion was again performed and the admission of dry air repeated. This exhaustion and admission of air were carried out alternately many times for the purpose of removing the comparatively large quantities of moisture which had been formed in the vessel during the process of making the various joints in the construction of the apparatus. When it was certain that the whole apparatus had been made fairly dry, the cylindrical electric heater was placed about the tube C, and while the exhaustion proceeded the tube was raised to a temperature of about 150° C., to hasten the removal of the gas present in large quantities in the pores of the charcoal at atmospheric pressure, and which separates from the charcoal rather slowly under reduced pressure if the temperature is kept low. When the mercury pump had been used to secure a fairly high vacuum the other parts of the apparatus, viz., the McLeod gauge, the viscosity apparatus, and the connecting tubes were heated to about 150° C., for the purpose of removing from the glass the occluded gases. After the pumping had proceeded for some time under these conditions, the heater was re-

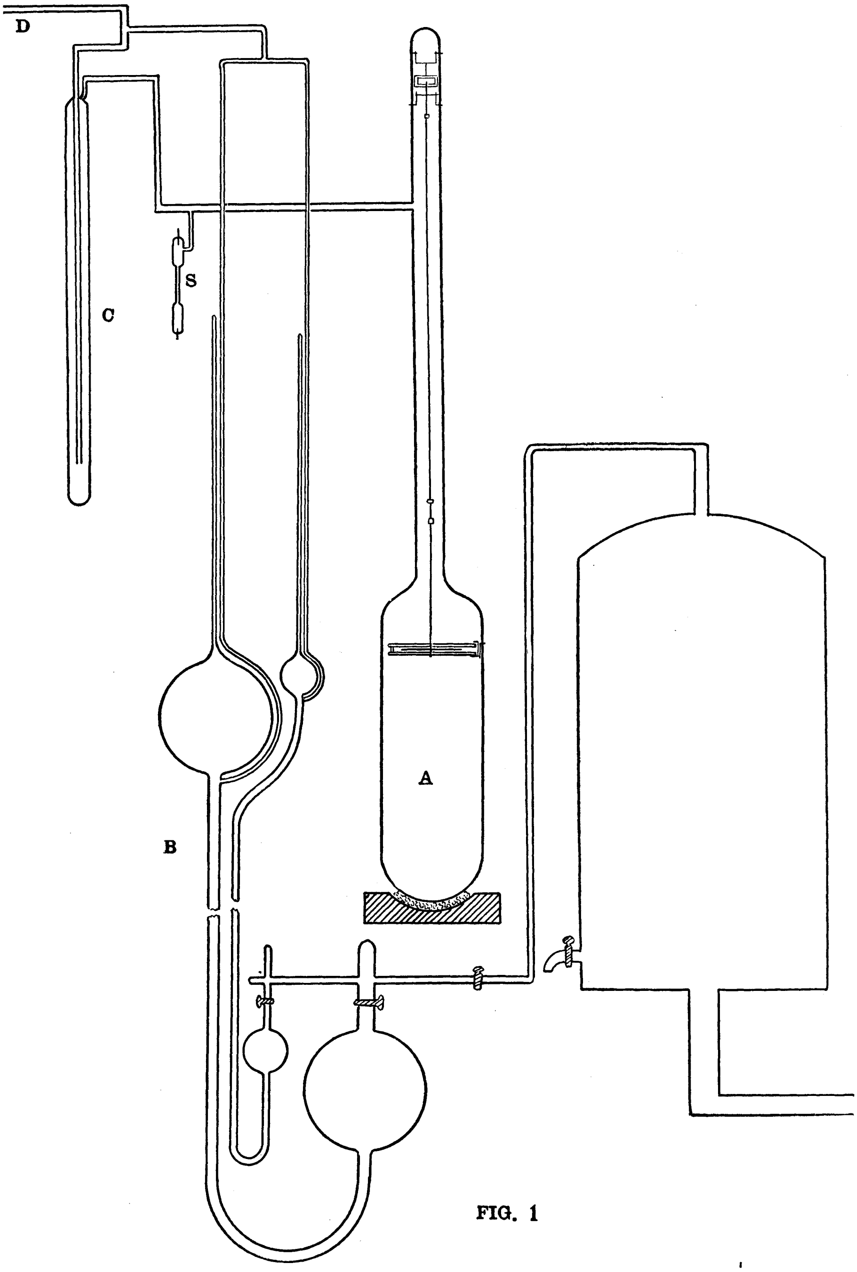

FIG. 1

moved from C and the Dewar vessel containing liquid air[3] was substituted for it, and the other part of the apparatus was allowed to cool down. The charcoal was allowed to absorb what it would at the temperature of the liquid air. Altogether the liquid air was kept surrounding the charcoal for about eighty hours, and from time to time during this interval a measurement of the decrement, l, was made. At first the diminution in the value of the decrement was fairly rapid, but after the first day the change was very slow. This, no doubt, was due in part to the slow passage of the gas towards the charcoal through the somewhat extended form of the apparatus. It was, also, probably due to the fact, which was noted later in the investigation, that at a given stage of exhaustion the raising of the free surface of the liquid air in the Dewar vessel surrounding C invariably produced a very appreciable diminution in the gas pressure in the apparatus, and the lowering of the free surface as the evaporation of the liquid air proceeded resulted in a distinct rise in the gas pressure. It is to be understood that the free surface was never allowed to fall as low as the top of the tube C, so that all of the charcoal was always below the free surface of the liquid air.

The following results show how the decrement changed with the time in the final forty-eight hours :

May 29,	12 M. to 2:53 A. M.	Decrement	0.000051
	7:15 P. M. to 8:58		0.000037
	10:53 P. M. to 1:36 A. M. (May 30)		0.000031
May 30,	1:36 A. M. to 4:48		0.000028
	11:11 A. M. to 2:06 P. M.		0.000037*
	2:06 P. M. to 5:27		0.000024*
	5:27 P. M. to 8:21		0.000028*
	8:21 P. M. to 11:49		0.000022

The smallest value of the decrement obtained was 0.000022, and this could be measured moderately well. Its error cannot, I think, be as much as ten per cent. Of course, it is clear that the true value of the decrement due to the friction in the fibre is somewhat less than this, for there is still, doubtless, some gas left to offer resistance to the moving disk, so that the number to be used for μ in the above equation should be somewhat smaller than 0.000022. I have ventured to make use of the value 0.000020 as the true value to which the decrement will approach as the exhaustion is pushed higher and higher. It will be seen from Table I that the calculations are carried through not only with this value, but also with the actually measured value 0.000022. This is done simply to show what effect such a change in the value of μ has on the series of results obtained.

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[3] The liquid air used in this investigation was obtained at the Chemical Laboratory, Harvard University.

* These were taken in the afternoon when there is considerable jarring of the apparatus and are probably not so accurate as the others.

It may be of interest to state that, at the stage of exhaustion when $\mu = 0.000022$ was obtained, the McLeod gauge indicated a pressure certainly less than 0.000001mm. It is, to be sure, of little value to give the measurement of a pressure by the gauge where a column of mercury a fraction of a millimeter high requires to be measured, and especially is this true where the tube containing the mercury has been heated and cooled repeatedly. The mercury has a habit of sticking to the glass to such an extent that pressure measurements under the conditions mentioned are surely not reliable. The value of the pressure given above, then, only indicates the order of magnitude of the pressure. Though the factor of the gauge used was 95813, yet it was quite inadequate to measure the pressure of the gas in the vessel.

Removal of Water Vapor and Mercury Vapor from the Hydrogen in the Viscosity Apparatus.

For this purpose it was necessary to make arrangements by which no vapor should be carried into the apparatus with the entering gas, and also all the vapor which was already in the apparatus might be taken out. The following arrangement was finally adopted, Figure 2. E is a U-tube of small bore, and bent so that it may enter the long Dewar vessel already mentioned. For reasons which will appear later it was found necessary for the remainder of the investigation to replace the tube C, Figure 1, by this tube E. F is a tube leading from the gas generator. It enters G, which is similar to C of Figure 1. It can be surrounded by a heater or a Dewar vessel as circumstances may require. A connecting tube leads from G to a point on the tube H, which connects E to the viscosity apparatus A, Figure 1. I leads to the pump and McLeod gauge. Anything which proceeds from the pump or McLeod gauge towards the viscosity apparatus must pass through E. Moreover, the gas entering from the generator will, with the given arrangement, retard the diffusion of mercury vapor from the pump and gauge towards the viscosity apparatus. If there is no vapor entering with the gas, there can be none entering the viscosity apparatus without passing through E, and, since throughout the experiment this tube was kept surrounded by the liquid air, the pressure of the vapor due to diffusion from the mercury in the pump or gauge could never exceed the vapor pressure of mercury at the temperature of the liquid air. The tube G, when surrounded with the liquid air, was sufficient safeguard against the entrance of water vapor with the gas.

The method of removing all water vapor and mercury vapor already in the apparatus beyond the tube E was that of repeated exhaustion and filling with the gas to be experimented with, the whole apparatus meanwhile being kept at a high temperature.[4]

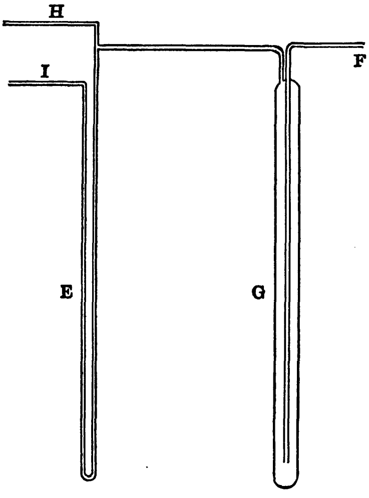

FIG. 2

At the first exhaustion, when the pressure had been reduced to a few centimeters of mercury, the tube G was surrounded by the electric heater, and the heat was applied to the oven in which the viscosity apparatus is placed. Practically the whole apparatus, except the gas generator, was kept hot while the pumping proceeded. After a fair vacuum was reached the pump was stopped and the hydrogen from the generator was allowed to enter very slowly, passing first over phosphoric pentoxide, and then over spongy platinum, heated in a combustion tube, before entering the tube G. This filling process was followed by another exhaustion under the same conditions. After the apparatus had been exhausted and filled a number of times in this way, when it seemed certain that the apparatus and the pores of the charcoal were filled with fairly pure hydrogen, the heater was removed from G, and the vessel containing the liquid air substituted for it. The same process of alternately exhausting and filling was continued, great care being taken in filling to allow the hydrogen to pass very slowly so that the drying process might be complete. Keeping the apparatus at a temperature of about 150° C. served to promote the evaporation of the mercury, which in all probability adhered to the inner glass surfaces. Comparatively large quantities of pure dry hydrogen were allowed to pass into the vessel and were then taken out. Each exhaustion would assuredly sweep out some vapor if it was present.

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[4] The suspended disk was, of course, lowered before this operation began.

It would naturally collect at E. We shall have evidence as to this later. After some days of incessant work the expected result was attained, as the character of the spectrum, obtained from the spectrum tube, S, showed. Even when the temperature of the viscosity apparatus was 150° C. the mercury lines were absent. The apparatus was then very slowly filled with hydrogen. The glass tube connecting G and H was then sealed off so that there were left no stop-cock joints to give trouble by leaking.

The Dewar vessel was removed from G, but the one surrounding E still remained. After the apparatus had cooled down to room temperature the disk of the viscosity apparatus was raised and adjusted as described in the former paper.[5]

METHOD OF EXPERIMENT.

The investigation of the relation of friction to pressure consists in measuring, for a given density of the gas, the logarithmic decrement of the suspended disk which is made to oscillate as a torsion pendulum between the two fixed plates of the apparatus.[6] The method of procedure was to measure the gas pressure in the apparatus by means of a manometer when the pressure was large, and by the McLeod gauge when it was small, and then to set the disk of the apparatus swinging and measure the decrement. Since the latter can be shown to be proportional to the resistance experienced by the disk, one gets data for the determination of the relation between friction and pressure.

It may be of interest to state here that in the first arrangement of the apparatus for the determination of the above relation instead of the simple bent tube E, a tube containing charcoal, similar to the tube C, was used. With this arrangement the mercury vapor was removed, but when observations on the decrement at different pressures were undertaken a difficulty presented itself. Although all of the tube containing the charcoal was immersed in the liquid air, the surface of which was always several inches above the top of the charcoal, yet it was found impossible to obtain a steady condition. As the evaporation of the liquid air proceeded, sufficient gas was given off from the charcoal to produce a large increase in pressure; as much as thirty per cent was observed. When a fresh supply of the liquid air was added the pressure diminished again. The difficulty became more serious as the pressure at which the observations were made became smaller.

The phenomenon was probably due to the fact that the fresh supply of liquid air was richer in nitrogen than it was after the process of boiling had proceeded for some hours. The nitrogen is the more volatile, and so the boiling will proceed more vigorously just after a fresh supply of air has been added than at any other time. Consequently the temperature of the boiling liquid will be lower at first than it is later, and the charcoal will thus absorb better at each addition of liquid air to the Dewar vessel. The charcoal is necessary for the phenomenon, for when the tube E was substituting for the tube containing the charcoal, the effect disappeared, or became inappreciable.

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[5] See pp. 133, 134.

[6] See pp. 124, 125 of former paper.

It was suggested earlier in the paper that there would be adduced evidence to show that the mercury driven out from the apparatus collected in the tube E. After the measurement of pressure and decrement had proceeded down to the least value given in the table, the supply of liquid air in the Dewar vessel in which E was placed was allowed to disappear gradually. As the evaporation proceeded, it was found that the decrement increased much more rapidly than the pressure as indicated by the McLeod gauge, showing that vapor was finding its way into the apparatus.

RESULTS.

In the first and second columns of Table I are contained the corresponding values of the decrement and pressure. Not all the numbers given in these columns were obtained by actual measurement. Only those which are marked with an asterisk were obtained in this way. The others were obtained as follows: A curve was plotted using the values of the pressure which were measured as abscissas and the corresponding values of the decrement as ordinates. This curve was drawn on such a scale that the value of the decrement, corresponding to any arbitrarily chosen pressure, could be obtained from the curve as accurately as it could be measured by the apparatus. The unmarked numbers in the first two columns were obtained by choosing arbitrarily a pressure and reading off from the curve the corresponding value of the decrement. In no case has this procedure involved an extrapolation.

After failing to obtain an analytical expression for the relation between the logarithmic decrement and the pressure which would be applicable over the whole range extending from very small pressures right up to atmospheric pressure, it was decided to find, if possible, an expression which would be applicable up to a certain pressure within the range for which it is known that the McLeod gauge measurements are quite reliable. Rayleigh[7] has shown that Boyle's Law holds down to 0.01mm. of mercury, and Baly and Ramsay[8] found the McLeod gauge measurements reliable for hydrogen. Recently Scheel and Heuse[9] have applied a membrane manometer, devised by them, to test Boyle's Law, and McLeod gauge measurements for air, and they state that they find both valid down to about 0.0001 mm., provided proper care is taken in drying the gas.

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[7] Phil. Trans., 196 (1901).

[8] Phil. Mgg., 38 (1894).

An examination of the results at pressures less than 0.1 mm. showed that the equation given above, viz.:

$$\left(\frac{k}{l-\mu}-1\right)p=c,$$

served the purpose exceedingly well, if the experimentally determined value of μ given above was used, and if the constants c and k were determined by means of values of p between 0.1 mm. and 0.01 mm. of mercury, and the corresponding values of l.

The pressures chosen for the determination of these constants were 0.0239 mm. and 0.0150 mm., the former of these being a pressure actually measured by the gauge, while the latter was chosen arbitrarily. The measurement of l corresponding to the former was 0.00936, while the value of l corresponding to the latter was 0.00620, and was obtained from the curve as described above. The values of the constants calculated from the above data are

$$c = 0.1491$$
$$k = 0.0676.$$

The equation now takes the form

$$\left(\frac{0.0676}{l-0.000020}-1\right)p=0.1491.$$

How well the equation gives the relation existing between p and l can be seen by a comparison of the numbers in the second and third columns of the table. A number in the third column is obtained by choosing a value of l from the first column, inserting it in the equation and deducing the value of p, which is then placed in the third column in the same horizontal row as the chosen value of l. It will be seen that the various numbers in this column agree very well with the corresponding numbers in the second column, except at the very lowest pressure, 0.000016 mm. used, when the difference is about twenty-five per cent.

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[9] Verhandl. d. Deutsch. Physikal. Gesellsch., **11**, 1 (1909).

If instead of using the value 0.000020 for μ, we make use of 0.000022, which was the smallest value of the decrement actually measured, the values of c and k are

$$c = 0.1494$$
$$k = 0.0677$$

and the fourth column gives the values of p calculated, in the way described, from the equation with these values for the constants instead of those used in the preceding case. This calculation is carried out to call attention to the magnitude of the change produced by a slight change in the value of the constant, μ, which is subject to some uncertainty, as has been shown. It will be seen that it is only where the decrement, l, is very small that the difference between the two results is appreciable. The smallest value of p in the fourth column is nearer to the corresponding value of p, measured by the McLeod gauge; but the measured value is subject to an inaccuracy about as great as the difference between the measured and calculated values of p.

The results given above make it highly probable that the measurements of pressure by the McLeod gauge are reliable in the case of pure, dry hydrogen for pressures as low as the smallest pressure recorded in the table.

It is to be observed that for pressures below, say, 0.01 mm. of mercury the friction with which we have to do is largely external friction, and this is proportional to the density of the gas and the mean molecular speed. The friction, and, therefore, the decrement, corresponding to a given pressure will be smaller for hydrogen than for, say, oxygen, or mercury vapor. In the case of mercury vapor the decrement at a given low pressure ought to be about ten times as great as it is for hydrogen at the same pressure, since the molecular weight of mercury is about one hundred times that of hydrogen, while the mean molecular speed is about one-tenth as great as it is for hydrogen.

To be sure it does not follow that the decrement of a mixture of hydrogen and mercury vapor, in such proportions that the partial pressures of the two are the same, is simply the sum of the two decrements obtained when the gas and vapor are separate. If one accepts the expression deduced by Meyer[10] for the external friction of a gas, and applies the same method in considering external friction of mixtures as he does in dealing with the internal friction of mixtures, he will be able better to understand how the external friction of a mixture of gases depends upon the proportion in which the gases are mixed. Meyer shows that the coefficient of external friction is given by

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[10] Kinetic Theory of Gases, p. 210 (Eng. Trans.).

$$\tfrac{1}{4}\beta m N \Omega,$$

where m is the molecular weight of the gas; N is the number of molecules per unit volume; Ω is the mean molecular speed; and β is a constant depending upon the solid surface. He gives some experimental evidence to show that β is independent of the gas.

In the case of a mixture of gases where there are N_1 molecules of one kind and N_2 molecules of another, in each unit volume we have, if N is the total number of molecules in the unit volume,

$$N = N_1 + N_2,$$

and the mean molecular weight is given by

$$m = \frac{N_1 m_1 + N_2 m_2}{N}$$

where m_1 and m_2 are the molecular weights of the two gases mixed.

Since the temperatures of the two gases are the same,

$$m_1 \Omega_1{}^2 = m_2 \Omega_2{}^2 = m \Omega^2.$$

Therefore,

$$m\Omega = m_1 \Omega_1 \sqrt{\frac{N_1}{N} + \frac{m_2}{m_1} \cdot \frac{N_2}{N}}.$$

If Boyle's Law holds, which seems a fair inference from the results given above, then we may write

$$m\Omega = m_1 \Omega_1 \sqrt{\frac{p_1}{p} + \frac{m_2}{m_1} \cdot \frac{p_2}{p}},$$

where p_1 and p_2 are the partial pressures and p is the whole pressure under the given conditions. If β is independent of the nature of the gas it follows that the ratio of the external friction of the mixture to the external friction of the gas whose partial pressure is p_1, if it were in the vessel alone, is

$$\frac{N m \Omega}{N_1 m_1 \Omega_1} = \frac{N m_1 \Omega_1 \sqrt{\frac{p_1}{p} + \frac{m_2}{m_1} \cdot \frac{p_2}{p}}}{N_1 m_1 \Omega_1} = \frac{N}{N_1} \sqrt{\frac{p_1}{p} + \frac{m_2}{m_1} \cdot \frac{p_2}{p}}.$$

If the mixture is one of hydrogen and mercury vapor such that $p_1 = p_2$, the above ratio becomes about 14. This means that if the pressure is measured by the McLeod gauge, which takes no account of the mercury vapor, the friction of the mixture would be about fourteen times as much as it would be with the same hydrogen pressure as in this case, but with the mercury vapor absent. If $p_1 = 1000p_2$, the ratio is about 1.05, and if $p_1 = 10{,}000p_2$, it becomes about 1.005.

It might be urged with regard to the method described above for freeing the hydrogen from mercury vapor that the lowest pressure of vapor obtainable by the method used is the pressure of mercury vapor at the temperature of liquid air boiling at atmospheric pressure. This pressure at 0° C. is about 0.0005 mm., but what it is at the lower temperature mentioned can hardly even be conjectured. We have simply to fall back upon the spectroscopic test. The above discussion shows, however, that if this pressure is less than 0.001 of the pressure of the hydrogen it will not very seriously affect the results. If it is as low as 0.0001 of the hydrogen pressure, then the error in the observations will easily be greater than any error introduced in this way. Considering the lowest pressure reached, namely, 0.000016 mm., the vapor pressure of mercury at the temperature of liquid air, boiling under atmospheric pressure, would require to be as low as 0.000,000,016 in order that the ratio of the partial pressures should be 1:1000.

This case serves to show how important it may be to consider mercury vapor when we are dealing with these very low pressures. It indicates that, in all high vacua work where we are considering the properties of a particular gas, it is important that great care should be taken to exclude this vapor. The McLeod gauge, of course, takes no cognizance of it, and in fact serves to introduce the vapor where it is not wanted. In all cases where the vacuum is high, and it is desirable to know the pressure in the vessel, and yet keep the gas pure, it would be desirable to have a gauge which would not introduce any impurity.

If the inference made above as to the validity of the McLeod gauge measurements on gas pressure is allowed, then we can say that reliance may be placed upon the measurements of pressure from decrement measurements in the apparatus used in this investigation. This method need introduce no mercury vapor, but it takes account of all that is in the vessel. Moreover, a discussion similar to that used for mercury vapor will show that in the case of oxygen the decrement, corresponding to a certain gas pressure, will be about four times as great as it is in the case of hydrogen. In the case of oxygen, therefore, a pressure of 0.00001 mm. should be measured with an accuracy of from five to ten per cent. This would indicate an absolute error of less than 0.000,001 mm.

The investigation is now being extended to the case of oxygen and nitrogen. The data obtained by using these gases, besides showing whether their behavior is like that of hydrogen, should give some more information regarding the quantity, β, which enters the foregoing discussion.

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